

A 2-D numerical research on spatial variability of concrete carbonation depth at meso-scale

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Abstract. This paper discusses the spatial variability of the carbonation depth caused by the mesoscopic structure of the concrete and the influence of the spatial variability on the thickness of the concrete cover. To conduct the research, a method to generate the random aggregate structure (RAS) based on polygonal particles and a simplified numerical model of the concrete carbonation at meso-scale are firstly developed. Based on the method and model, the effect of the aggregate properties including shape, content and gradation on the spatial variability of the carbonation depth is comprehensively studied. The results show that a larger degree of the spatial variability will be obtained by using (1) the aggregates with a larger aspect ratio; (2) a larger aggregate content; (3) the gradation which has more large particles. The proper sample size and model size used in the analysis are also studied. Finally, a case study is conducted to demonstrate the influence of the spatial variability of the carbonation depth on the proper thickness of the concrete cover. The research in this paper not only provides suggestions on how to decrease the spatial variability, but also proposes the method to consider the effect of the spatial variability in designing the thickness of the concrete cover.

Keywords: concrete structures; carbonation; spatial variability; meso-scale

1. Introduction

Steel corrosion is one of the major problems which most reinforced concrete (RC) structures may encounter during their service life. In atmospheric environment where the concentration of the chloride ion remains in a very low stage, the steel corrosion process is mainly induced by the concrete carbonation process. In the fresh concrete, the pH of the pore solution can reach as high as 13.0 due to a large amount of alkaline products such as $\text{Ca}(\text{OH})_2$ and C-S-H, which are formed during the cement hydration process. In this alkaline environment, the steel rebars can be effectively protected and prevented from corroding. However, once the fresh concrete is exposed to the atmospheric environment, CO_2 will penetrate into the concrete and consume the alkaline substances. As a result, the pH of the pore solution continuously decreases. When the pH drops to a threshold value, the passivation layer on the surface of the steel rebar will become unstable and

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be dissolved, which eventually results in the initiation of the steel corrosion process.

To predict the concrete carbonation process, a lot of experimental, theoretical and numerical researches have been conducted during the past 40 years. As early as in 1989, Papadakis *et al.* (1989) already theoretically proved that the carbonation depth has an approximate linear relationship with the square root of the time, i.e., $d = k\sqrt{t}$. This linear model was preferred due to its simplicity and has been widely used in practical engineering. However, some complicated situations, e.g., multidimensional carbonation process, climate change, heterogeneity of composite materials, influence of rehabilitations, cannot be easily considered in this model. As a result, some numerical simulation researches were also conducted. Saetta *et al.* (1995) developed a 2-D model for carbonation and moisture/heat flow in porous materials and applied the model to simulate the multidimensional carbonation process, e.g., at the corner of a concrete structure. Burkan Isgor and Razaqpur (2004) developed a 2-D Windows-based finite element computer program based on a coupled model of heat transfer, moisture transport and concrete carbonation. One of the highlights in their research is that they used this model to study the carbonation processes in sound and cracked concrete. Park (2008) constructed a diffusion-reaction carbonation model using the finite element method, and applied the model to study the carbonation process in coated concrete. Their research demonstrated the convenience of the numerical method in considering the heterogeneity of composite materials, as well as the effect of rehabilitations, e.g., attaching special coatings to the surface of the concrete to prevent the penetration of CO_2 . Meier and Peter did a series of theoretical and mathematical research on the concrete carbonation (Meier *et al.* 2007a, Meier *et al.* 2007b, Peter *et al.* 2008). In their researches, a new complicated numerical model of the concrete carbonation was established. By using this model, Peter *et al.* (2008) studied the competition of several carbonation reactions in concrete by 1-D numerical method. According to their research, the reactions between CO_2 and some unhydrated substances such as C_3S and C_2S can be neglected and also the C-S-H has a quite limited effect on the overall carbonation process. Other numerical simulation researches can be also found somewhere else (Papadakis *et al.* 1991b, Bary and Sellier 2004, Wang and Lee 2009, Radu *et al.* 2013).

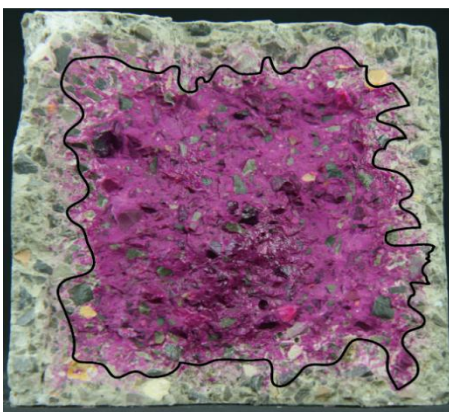


Fig. 1 Identification of carbonation depth by phenolphthalein in real concrete specimen

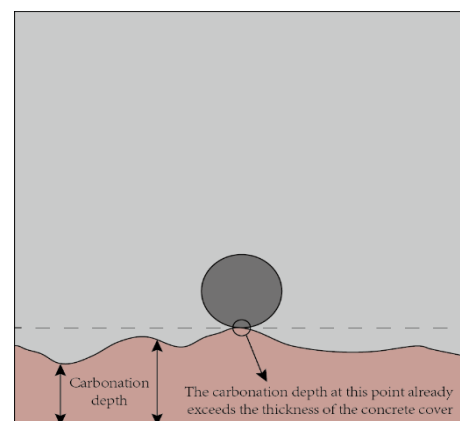


Fig. 2 A schematic of the spatial variability of concrete carbonation depth

As a summary, most of the above numerical researches mainly focus on how to predict the carbonation depth from an average point of view. A typical common characteristic of these researches is that concrete is often assumed as a homogeneous material. With this assumption, the carbonation front will propagate into concrete uniformly. Therefore, the shape of the carbonation front should be a straight line. However, this result is quite different from the experimental observations. Fig. 1 shows an example of identifying the carbonation depth by phenolphthalein in a real concrete specimen. It is obvious that the identified carbonation front is not a straight line parallel to the exposed surface, but a curve with different carbonation depths at different positions. This phenomenon confirms the spatial variability of the carbonation depth at meso-scale. Since the specimen is exposed to a homogeneous environmental condition in the carbonation chamber, this spatial variability should be mainly attributed to the heterogeneous nature of the concrete itself.

The above spatial variability of the carbonation depth has a potential influence on the prediction on the initiation time of steel corrosion caused by the concrete carbonation. Fig. 2 shows a schematic of this issue. While the carbonation depths in the other positions remain in a low level, the carbonation depth near the position of the steel rebar may already exceed the thickness of the concrete cover. In this situation, the traditional concrete carbonation models based on the average carbonation depth may underestimate the initiation time of steel corrosion, which will result in an unsafe prediction. This problem also exists in the chlorine-induced steel corrosion, which was discussed in a recently published paper (Angst and Polder, 2014). Their experimental research confirmed the existence of the spatial variability of chloride concentration in concrete. In their opinion, the primary cause should be identified to be the presence of coarse aggregates, i.e., the mesoscopic structure of concrete. Inspired by this research, whether the mesoscopic structure of concrete has a similar effect on the concrete carbonation depth and to what degree the effect is should be comprehensively studied. Up to now, several researches on this topic have been already conducted. Ruan and Pan (2012) did a preliminary research on the mesoscopic carbonation process in the concrete. Their research qualitatively revealed the effect of the aggregates on the shape of the carbonation front. Huang *et al.* (2012) studied the effect of the coarse aggregates on the carbonation depth in a probabilistic way. In these researches, the shape of the aggregate was assumed as a circle or sphere which is over-simplified, and also the significance of the mesoscopic carbonation process in the design of the concrete cover was not discussed. Instead of explicitly modelling the mesoscopic structure of the concrete, the multi-scale approach were also used in some papers (Peter and Bohm 2009, Heine *et al.* 2013). The general idea of the multi-scale approach is to use the method of the so-called “homogenization” to create a homogeneous macroscopic model. To obtain the parameters used in the macroscopic model, some other numerical simulations are usually conducted at smaller scales, e.g., meso-scale or even micro-scale. Based on the results of these numerical simulations, a so-called “equivalent” parameter is determined and passed back to the macro-scale. This procedure is sometimes called the “parameter passing” (Qian 2012). In this way, although the final simulation is carried out at macro-scale, the parameters used in the simulation have already considered the characteristics of the material at smaller scales. Thus, the multi-scale approach can greatly increase the efficiency of the numerical simulation. However, since the final simulation is conducted in a homogenized material, the spatial variability of the concrete carbonation depth still cannot be explicitly reflected.

This paper is a continuous research following Ruan and Pan (2012) and Pan *et al.* (2014). The topic of the spatial variability of concrete carbonation depth is the main concern of this paper. To conduct the research, a new meso-scale model of concrete based on polygonal aggregates is firstly developed. The aspect ratio is chosen as the key parameter controlling the shape of the aggregates.

Secondly, a simplified numerical model of the concrete carbonation is provided. This model was also used in the previous research by Ruan and Pan (2012). Experimental data from published papers is adopted to verify the numerical model of the concrete carbonation. Thirdly, the effect of the aggregates, e.g., the gradation, the shape, the usage, on the spatial variability of the carbonation depth is comprehensively discussed in a statistical manner. Proper sample size and model size are also found based on the statistical analysis. The research in this paper may provide a new insight into the probability-based design method of the concrete cover.

2. Numerical models and methods

2.1 Meso-scale model of concrete based on polygonal aggregates

At meso-scale, concrete is usually considered as a composite material, where the aggregates are randomly distributed in the cement paste. Therefore, the main task of constructing the meso-scale model of concrete is to simulate the aggregates. In reality, the shape of aggregate is very complicated, especially in the case of crushed stones from rocks. If the mesoscopic analysis is performed on a real sample, e.g., performance evaluation of in-service structures, the aggregate shape can be obtained precisely by an image scan (Nagai *et al*, 2000; Huet, 1993). However, in other situations, such as in the preliminary structural design, no sample is available, and the

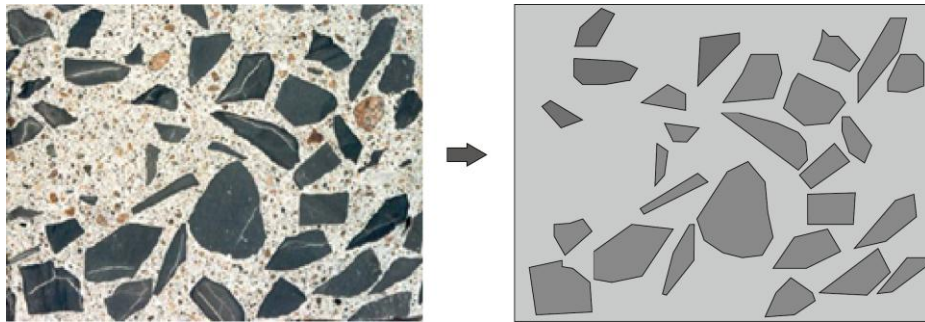


Fig. 3 Real coarse aggregates and approximation using polygons

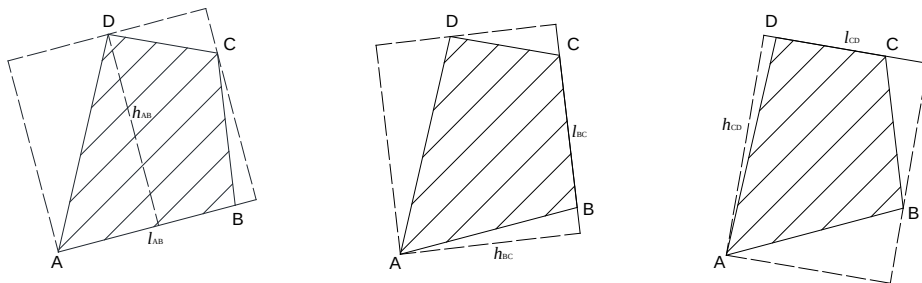


Fig. 4 Definition of bounding rectangle for arbitrary polygon

aggregate has to be simulated numerically. In such cases, approximations of the aggregate shape must be adopted. This paper simplifies the shape of the aggregate and assumes it is a polygon. This simplification is especially reasonable to simulate the shape of the coarse aggregates whose angularity cannot be neglected. Fig. 3 shows a section cut from a real concrete specimen and the corresponding approximation using polygons.

To describe how to generate the polygonal aggregates, the aspect ratio of an arbitrary polygon should be firstly defined. For the polygon shown in Fig. 4, the aspect ratio can be determined by the following procedure.

- (1) Choose a side of the polygon as the reference line, e.g., line AB;
- (2) Determine the bounding rectangle based on the reference line, as shown in Fig. 4;
- (3) Determine the length and height of the bounding rectangle, i.e., l_{AB} and h_{AB} ;
- (4) Choose the other sides of the polygon as the reference lines and repeat the above steps to obtain the corresponding l_i and h_i ;

- (5) The aspect ratio of the polygon can be defined as

$$\beta = \max(l_i/h_i) \quad (1)$$

To ensure that the generated polygonal aggregates can satisfy a given gradation, a basic circle is firstly generated and an initial polygon is cut from this circle. After that, this initial polygon is stretched and scaled to form the final polygon, as shown in Fig. 5. This procedure is to ensure that the aspect ratio of the final polygon is the same as the needed one and the area of the polygon is the same as that of the basic circle.

The separation check for polygonal aggregates can be simply conducted by determining whether all the vertices of a polygon are outside the boundaries of the other polygons and whether any edge of the polygon intersects the edges of the other polygons. After the position of a polygonal aggregate is determined, the information of this aggregate can be uniquely defined as $[(x_0, y_0), (x_1, y_1), (x_2, y_2) \dots (x_n, y_n)]$, where (x_i, y_i) is the global coordinates of the vertices of the polygon.

In practice, the content of the aggregate is usually expressed as the three-dimensional volume fraction, which should be converted to the two-dimensional area fraction, since only a two-dimensional analysis is conducted in this paper. According to the research by Zheng (2000), the area fraction should equal to the volume fraction, i.e., $A_a = V_v$. Although this conclusion is achieved based on the spherical particles, it is used here as an approximation.

Based on the above method, two typical examples of the meso-scale model based on the polygonal aggregates are shown in Fig. 6.

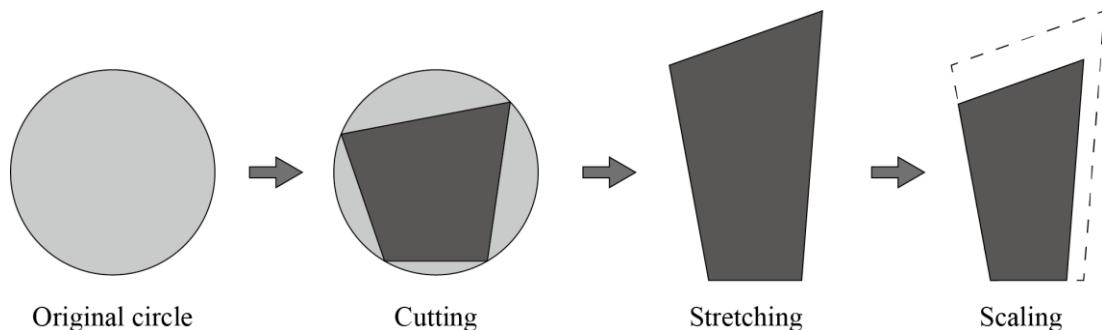


Fig. 5 Generation of polygonal aggregate from basic circle

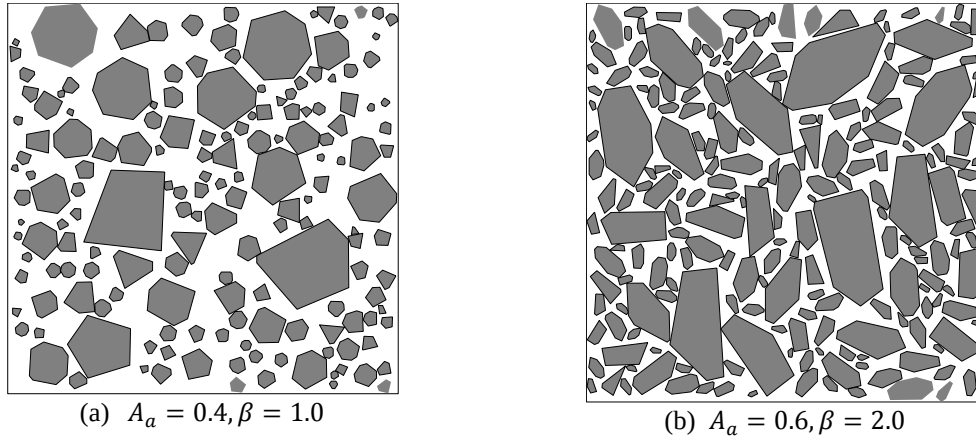


Fig. 1 Examples of 2-D meso-scale model based on polygonal aggregates

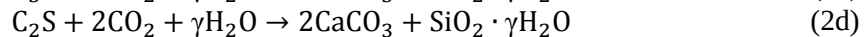
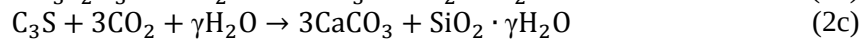
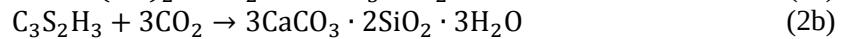
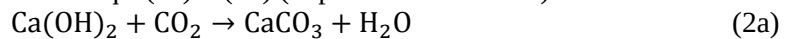
Table 1 Meanings of variables in Eq. (3)

Variable	Meaning	Unit
ϕ	porosity of cement paste	-
s	saturation degree	-
$D_{CO_2}^e$	effective diffusion coefficient of CO_2	m^2 / s
$f_{C,CH}$	rate of chemical reaction between CH and CO_2	$mol / (m^3 \cdot s)$ (molar per unit volume of pore air per second)
$[CH(s)]$	concentration of CH in solid state	mol / m^3 (molar per unit volume of cement paste)
$[CO_2(g)]$	concentration of CO_2 in gaseous state	mol / m^3 (molar per unit volume of pore air)

2.2 Numerical model of concrete carbonation

2.2.1 Governing equations

Generally, four chemical reactions exist during the carbonation process of Ordinary Portland Cement (OPC) concrete, as shown in Eqs. (2a) ~ (2d) (Papadakis *et al.* 1989).



In a well hydrated OPC concrete, the amount of the unhydrated substances, e.g., C_3S and C_2S , is very small. Therefore, the chemical reactions of these substances with CO_2 just have ignorable contributions to the overall concrete carbonation, and can be neglected as a result (Peter *et al.* 2008). The amount of C-S-H is comparable with that of $Ca(OH)_2$. However, the reaction rate of Eq. (2b) is much slower than that of Eq. (2a), which means that the existence of C-S-H cannot effectively prevent CO_2 from penetrating into the concrete (Papadakis *et al.* 1989). Therefore, the

chemical reaction between C-S-H and CO_2 is also neglected in this paper. It should be pointed out that this neglect overestimates the amount of CO_2 which reacts with $\text{Ca}(\text{OH})_2$, and will result in a safe prediction of the carbonation depth.

Based on the above simplifications, a numerical model of the concrete carbonation is developed. The governing equations of this model are shown in Eqs. (3a) and (3b). The meaning of each variable is listed in Table 1. The derivation process of Eqs. (3a) and (3b) can be found in Ruan and Pan (2012).

$$\partial_t\{\phi(1-s)[\text{CO}_2(\text{g})]\} = \text{div}\{D_{\text{CO}_2}^e \cdot \mathbf{grad}[\text{CO}_2(\text{g})]\} - \phi s f_{\text{C,CH}} \quad (3a)$$

$$\partial_t[\text{CH}(\text{s})] = -\phi s f_{\text{C,CH}} \quad (3b)$$

2.2.2 Effective diffusivity of CO_2

The concrete carbonation in reality is a fast reaction but slow diffusion process, which means the rate of the chemical reaction is much faster than the diffusion of CO_2 . Thus, the effective diffusion coefficient of CO_2 ($D_{\text{CO}_2}^e$) here is of special importance. In this paper, the model shown in Eq. (4) which is initially proposed by Papadakis *et al.* (1991a) is used.

$$D_{\text{CO}_2}^e \approx 1.64 \times 10^{-6} \phi_p^{1.8} (1 - RH)^{2.2} \quad (4)$$

where ϕ_p is the porosity of the cement paste. Its initial value before carbonation can be evaluated by Eq. (5) proposed by Hansen (1986).

$$\phi_p^0 = \frac{w/c - 0.17m}{2/c + 0.32} \quad (5)$$

where w/c is the water cement ratio, and m is the overall degree of the cement hydration.

As the reduction of CH and CO_2 produces CaCO_3 , the value of ϕ_p in Eq. (4) will decrease due to the increase in molecular weight. Park (2008) proposed an empirical model shown in Eq. (6) to consider the variation of porosity due to carbonation process.

$$\phi_p' = \lambda \phi_p^0 \quad (6)$$

where ϕ_p^0 is the porosity before carbonation, while ϕ_p' is the porosity after carbonation. $\lambda = 0.92 - 3.95 \cdot 0.94^{100 \cdot w/c}$ is the reduction coefficient.

Eq. (6) only provides the values of porosity at two typical time points: before and after carbonation. In order to get the value of porosity during the carbonation process, a linear interpolation is employed as follows

$$\phi_p(t) = [(\lambda - 1)\alpha_c(t) + 1] \cdot \phi_p^0 \quad (7)$$

where $\alpha_c(t) = 1 - [\text{CH}(\text{s})](t)/[\text{CH}(\text{s})]_0$ is the carbonation degree.

2.2.3 Water vapor sorption isotherms

In atmospherical environment, the water content is usually expressed in relative humidity. This parameter can be converted into saturation degree in Eqs. (3a) and (3b) by the Water Vapor Sorption Isotherms (WVSIs). Based on the data from a series of experiments by Baroghel-Bouny (2007), the following equation is used for data fitting. The result is shown in Fig. 7. During these data fittings, the value of a is calculated by Eqs. (9a) and (9b), while the value of b is a constant of -2.750 for adsorption and -1.750 for desorption processes.

$$y = ax(x^2 - 1) + bx(x - 1) + x \quad (8)$$

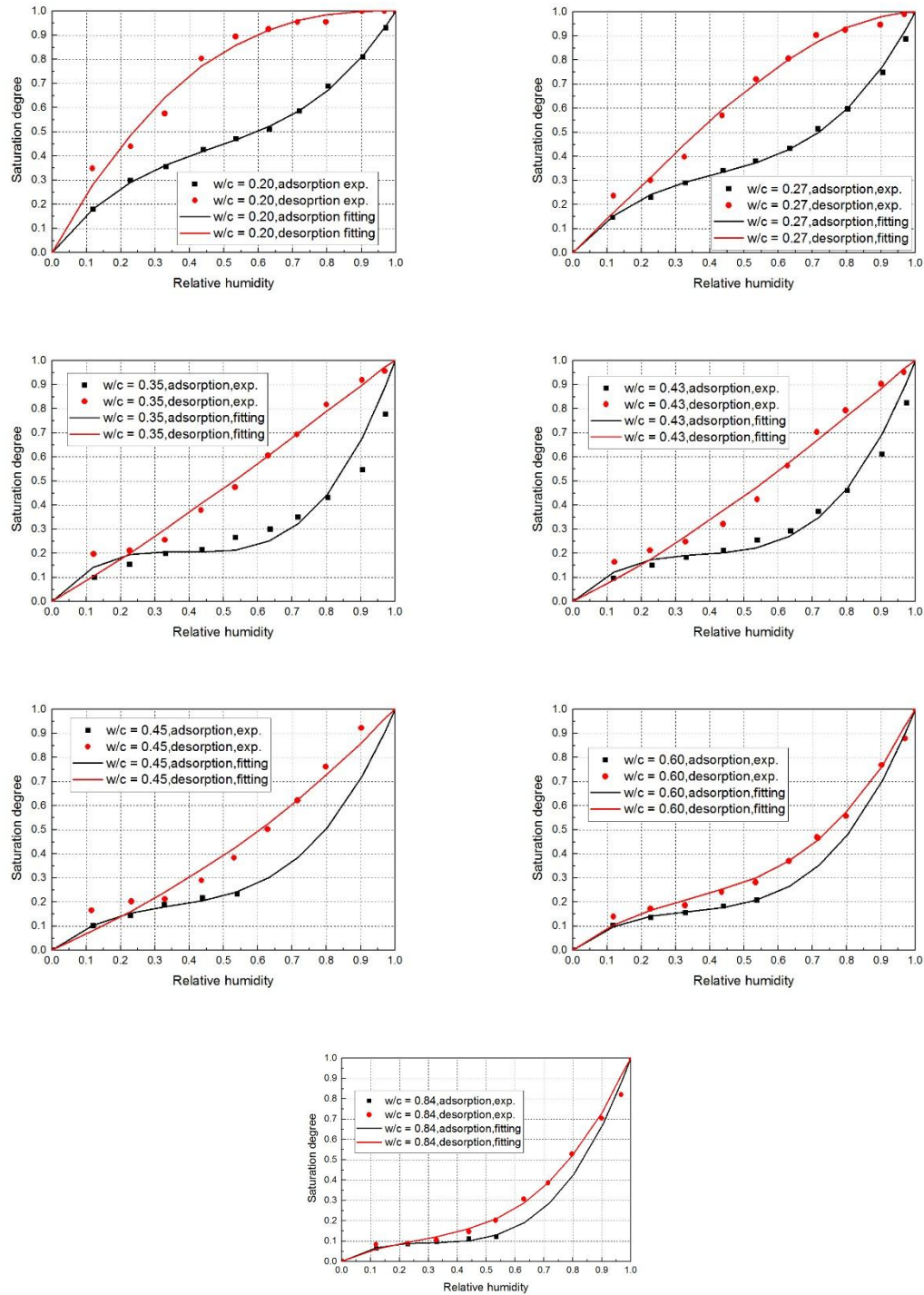


Fig. 7 Data fitting of WVSIs

$$\text{adsorption} \quad a = 0.5859 \ln(w/c) + 3.0285 \quad (9a)$$

$$\text{desorption} \quad a = 1.2016 \ln(w/c) + 2.3258 \quad (9b)$$

2.3 Finite element method

Once the meso-scale model of the concrete is obtained, this model is projected to a background mesh with regular triangle elements. The type of each element is determined according to the position of the triangle in the meso-scale model, as shown in Fig. 8. Three different types of elements, namely cement paste element, aggregate element, interfacial transition zone (ITZ) element, are defined in the final finite element model.

Based on some experimental researches (Ollivier *et al.* 1995, Prokopski and Halbiniak 2000), the ITZ is a special region in concrete in that it has a higher porosity than the cement paste. Therefore, the diffusivity of some chemical substances such as CO_2 in the ITZ is usually several times than that in the cement paste. It should be pointed out that the thickness of the ITZ used in the numerical simulation is usually increased on purpose. The reason for this is that the real

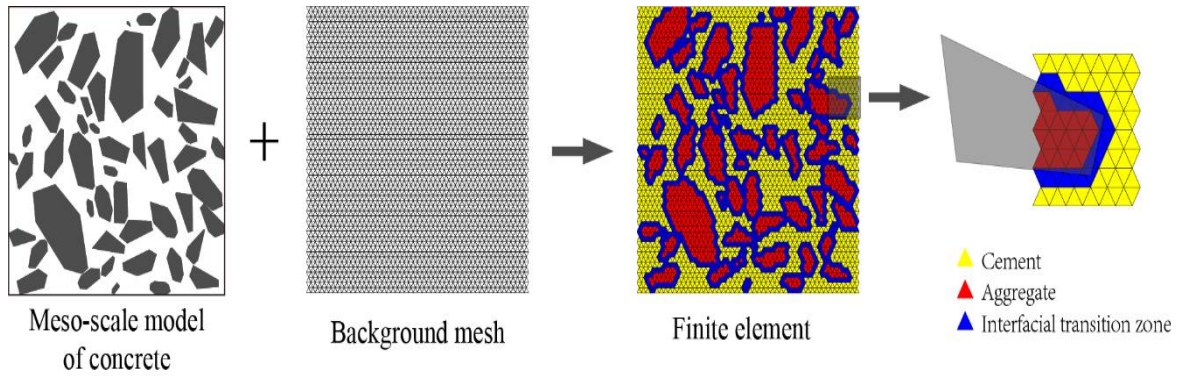


Fig. 8 Formation of finite element by background mesh

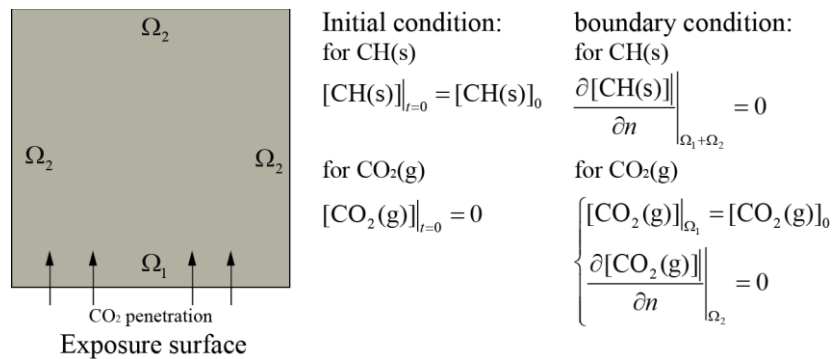


Fig. 9 Illustration of initial and boundary conditions commonly used in numerical simulation of concrete carbonation process

thickness of the ITZ observed in experiments is about 10~50 μm (Ollivier *et al.* 1995, Scrivener and Nematì 1996). If this tiny size is used as the length of the triangle element, which determines the thickness of the ITZ in the numerical model, an unacceptable amount of the computational time may be encountered. In this paper, the thickness of the ITZ is set as 0.3mm, and the diffusivity of CO_2 in the ITZ is assumed as 3.0 times than that in the cement paste. This result is obtained according to the “try and error” method based on the experimental data shown in Fig. 10. It should be pointed out that the diffusivity of CO_2 in the ITZ here is dependent on the mesh size. As a result, the value 3.0 here can only be used when the thickness of the ITZ is set as 0.3mm in the background mesh. Further research is obviously necessary to obtain a better method to model the ITZ in the meso-scale model of the concrete.

Once the meso-scale model is meshed, the partial differential equations, i.e., Eqs. (3a) and (3b), can then be numerically solved by the finite element method. The initial and boundary conditions used in most simulations are shown in Fig. 9.

Generally, the boundary conditions of the concrete carbonation process can be divided into two kinds, i.e., Ω_1 for exposure surface and Ω_2 for non-exposure surface. Ω_2 here may refer to the surface coated with the material such as epoxy which can prevent CO_2 from penetration, or the boundary inside the concrete. The initial concentration of $[\text{CH}(\text{s})]$ ($[\text{CH}(\text{s})]_0$) can be calculated according to the cement hydration process. However, this topic is beyond the scope of this paper. Detailed information can be found in Papadakis *et al.* (1991a). To avoid the complicated kinetics of the cement hydration, it is usually assumed that a fully hydration can be achieved before carbonation. Under this assumption, the value of $[\text{CH}(\text{s})]_0$ is usually in the order of $10^3 \text{mol} \cdot \text{m}^{-3}$ (in molar per unit volume of concrete). In practice, the concentration of gaseous CO_2 is usually expressed in the form of the volume fraction ($[\text{CO}_2(\text{g})]_v$), which should be changed by $[\text{CO}_2(\text{g})] = 22.4 \cdot 10^{-3} [\text{CO}_2(\text{g})]_v$ to the unit in Table 1. In this way, the boundary value of gaseous CO_2 ($[\text{CO}_2(\text{g})]_0$) can be calculated.

2.4 Experimental verification

To verify the numerical model of the concrete carbonation in this paper, the experimental data from Papadakis *et al.* (1991b) is used. In their research, the carbonation processes not only in the cement paste but also in the concrete specimens with different volume fractions of aggregates were investigated. Correspondingly, these carbonation processes are simulated here to make comparisons with the experimental results.

2.4.1 Experiment on pure cement paste

According to the description of the experiment in Papadakis *et al.* (1991b), the simulation of the carbonation process in the cement paste is conducted in a 2-D rectangle with the size of $0.1\text{m} \times 0.1\text{m}$. The water cement ratio used in the experiment is 0.50. The volume concentration of CO_2 and the relative humidity in the carbonation chamber are 50% and 65%, respectively. The values of $[\text{CH}(\text{s})]_0 = 1950 \text{mol}/\text{m}^3$ and $[\text{CO}_2(\text{g})]_0 = 22.32 \text{mol}/\text{m}^3$ in Fig. 9 are calculated for the initial and boundary conditions. Since the carbonation depth in the experiment was measured by the phenolphthalein which will change the color when $\text{pH} = 8.0 \sim 10.0$, the criterion of $\text{pH} = 8.0$ is used to identify the carbonation front in the simulation. A comparison of the simulation results with the experimental results is shown in Fig. 10. As can be seen, the numerical simulation reproduces the experimental results well.

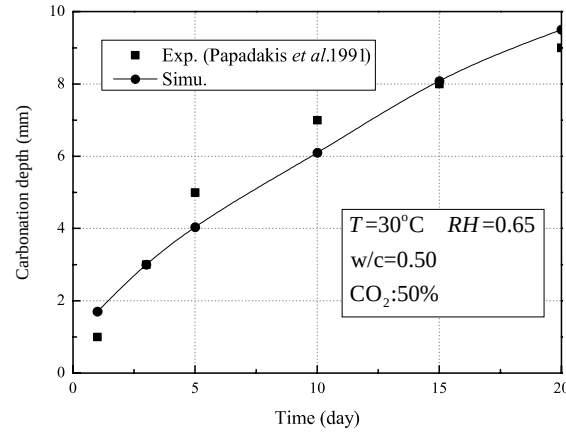


Fig. 10 Experimental verification on cement paste

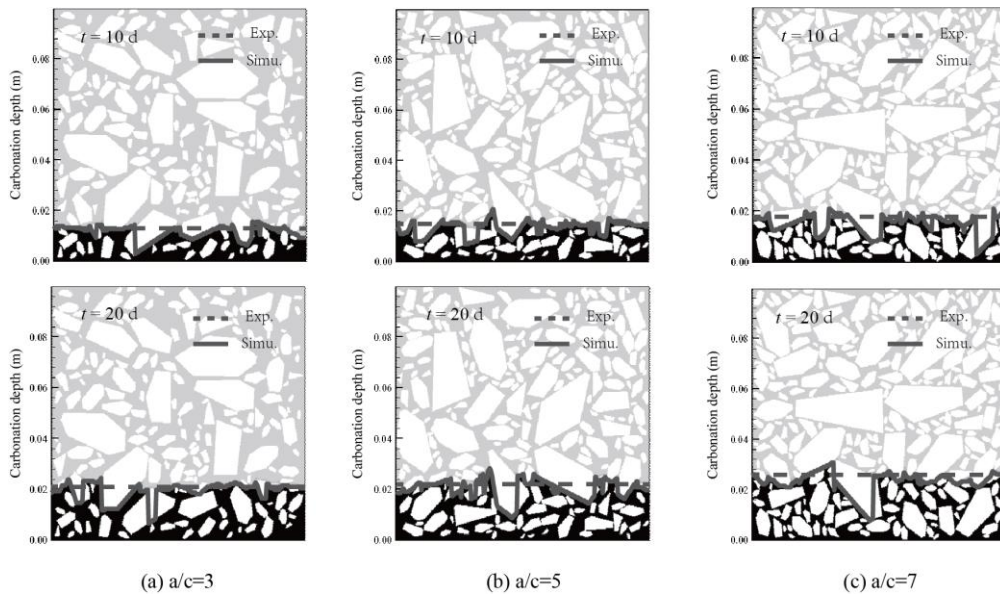


Fig. 11 Experimental verification on concrete

2.4.2 Experiment on concrete

Three different concrete specimens with different volume fractions of aggregates were cast in the experiment by Papadakis *et al.* (1991b). The aggregate gradation used in the experiment is shown in Fig. 19 (labelled as Type I). The water cement ratio (w/c) of the concrete specimens is 0.65. The environmental conditions in the carbonation chamber are the same as those in the experiment on the cement paste. The aggregate cement ratios (a/c) of these three concrete specimens are 3.0, 5.0 and 7.0, respectively. Based on the mix proportion of the concrete, these

aggregate cement ratios can be converted to the volume fractions of aggregates which are 0.544, 0.665 and 0.736, respectively. For the initial and boundary conditions shown in Fig. 9, $[\text{CH}(\text{s})]_0 = 1650 \text{ mol/m}^3$ and $[\text{CO}_2(\text{g})]_0 = 22.32 \text{ mol/m}^3$ are calculated. The numerical and experimental results in the three concrete specimens are shown in Fig. 11(a), Fig. 11(b) and Fig. 11(c), respectively. According to these comparisons, the numerical model of the concrete carbonation in this paper can be verified.

3. Spatial variability of concrete carbonation depth at meso-scale

3.1 Definition

Before quantitatively discussing the spatial variability of the carbonation depth, a mathematical definition to describe the exact meaning of the spatial variability should be given. As mentioned in the Introduction of this paper, when the concrete carbonation process is simulated at meso-scale, the carbonation front is not a straight line parallel to the exposure surface, but a curve distorted by the aggregates. Therefore, the mesoscopic carbonation depth should be a function of the position where the depth is measured. In numerical simulation, since finite element method (FEM) is used, the entire model is discretized. Therefore, the above function should be also in a discrete form, which leads to

$$d_i(t) = f(x_i(t)) \quad i = 1, 2, 3 \dots N \quad (10)$$

where $d_i(t)$ is the carbonation depth at the position of x_i , and at the time of t . x_i is the position on the exposure surface. N is the total number of the nodes on the exposure surface in the finite element mesh.

The standard deviation (SD) of $d_i(t)$ can be calculated as

$$\sigma_d(t) = \sqrt{\frac{1}{N} \sum_{i=1}^N [d_i(t) - \mu_d(t)]^2} \quad (11)$$

where

$$\mu_d(t) = \frac{1}{N} \sum_{i=1}^N d_i(t) \quad (12)$$

σ_d in Eq. (11) can be seen as the major parameter which represents the degree of the spatial variability of the carbonation depth.

In practical engineering, it may be more important that a confidence interval of the carbonation depth, i.e., $\mu_d \pm \alpha \cdot \sigma_d$, can be given to design the thickness of the concrete cover. It is well known that for a normal distribution $X \sim N(\mu, \sigma^2)$, the values of α for 68.2% and 95.4% confidence levels are 1.0 and 2.0, respectively. However, for the random variable in discrete form such as $d_i(t)$ defined in Eq. (10), the value of α should be determined according to the following procedure.

- (1) Determine the expected confidence level ε ;
- (2) Set initial values: $\alpha(t) = \alpha_0$ and $n = 0$, where α_0 is the first guess of α , n is a counter;
- (3) For each position $x_i(t)$ on the carbonation front, determine the corresponding carbonation depth $d_i(t)$. If $\mu_d(t) - \alpha(t) \cdot \sigma_d(t) \leq d_i(t) \leq \mu_d(t) + \alpha(t) \cdot \sigma_d(t)$, let $n = n + 1$;
- (4) If $n/N \geq \varepsilon$, go to Step (5). Otherwise, let $\alpha(t) = \alpha(t) + \delta\alpha$ and $n = 0$, then go to Step (3). $\delta\alpha$ refers to the increment of α , which should be small enough to ensure the accuracy of α .
- (5) Record the current value of α . Choose another time t and go back to Step (2) until all the time points are dealt with.

Based on the above definitions, the effect of the spatial variability of the carbonation depth on the thickness of the concrete cover can be defined. In a traditional macroscopic approach, the thickness of the concrete cover is determined from an average point of view, which means.

$$D_{c,1} = \mu_d(T) \quad (13)$$

where T is the design service life of the concrete component.

After the spatial variability of the carbonation depth is considered, the thickness of the concrete cover should be determined according to the confidence interval, as shown in the following equation.

$$D_{c,2} = \mu_d(T) + \alpha(T) \cdot \sigma_d(T) = \mu_d(T)[1 + \alpha(T) \cdot \delta_d(T)] = \gamma(T) \cdot \mu_d(T) \quad (14)$$

where the coefficient of variation (CV) δ_d is defined as

$$\delta_d(t) = \frac{\sigma_d(t)}{\mu_d(t)} \quad (15)$$

By comparing Eq. (13) with Eq. (14), one may find that $\gamma(T)$ can be seen as a partial factor which represents the uncertainty caused by the spatial variability of the carbonation depth at meso-scale. Based on Eqs. (13) and (14), the difference between the thicknesses of the concrete cover with and without the consideration of the spatial variability of the carbonation depth can be calculated as

$$\Delta D_c = [\gamma(T) - 1.0] \cdot \mu_d(T) = \alpha(T) \cdot \sigma_d(T) \quad (16)$$

3.2 Preliminary case study

Based on Eqs. (11) ~ (16), a typical case study is performed. In this case study, the area fraction, aspect ratio and gradation type of the aggregates are 0.5, 1.0 and Type I (see Fig. 19), respectively. The other material and environmental parameters are kept the same as those in the experiment mentioned in Section 2.4.2. This case study can be seen as a virtual accelerated carbonation experiment. In mesoscopic research, the main advantage of the virtual experiment is that once the numerical model used in the virtual experiment is verified, a large number of samples can be

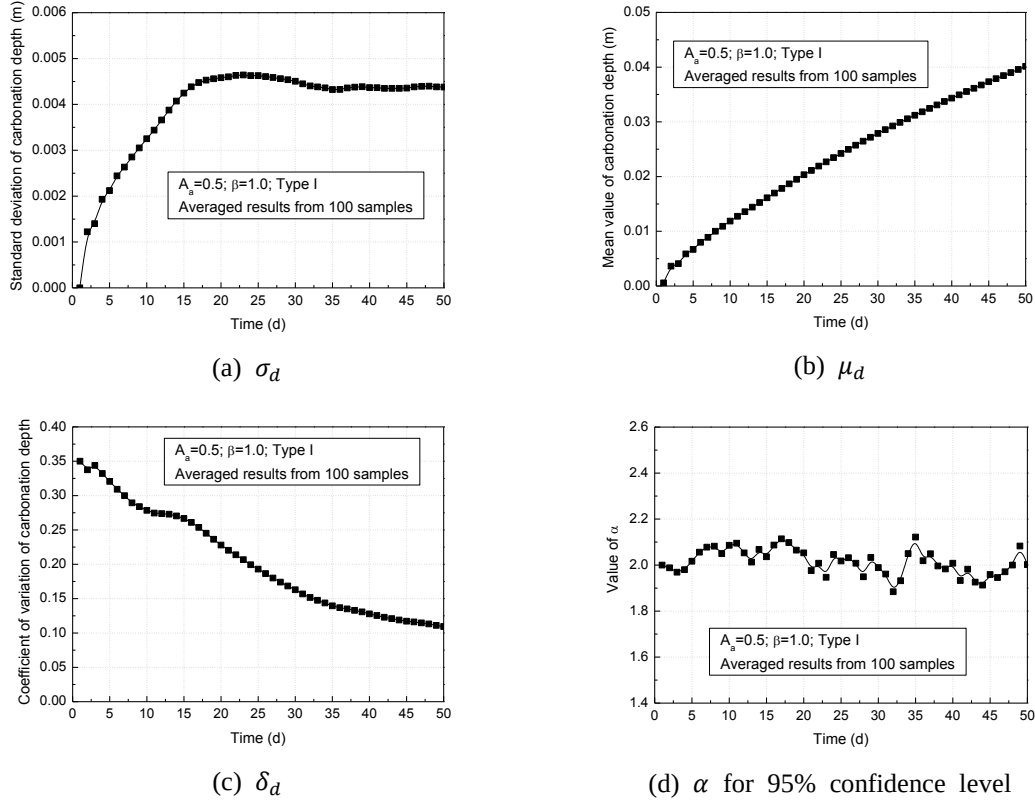


Fig. 12 A typical case study on spatial variability of carbonation depth

numerically generated and a statistical analysis can be easily conducted. However, this kind of statistical analysis is difficult to perform in a real experiment due to the unacceptable cost and time.

Since the meso-scale model is randomly generated, 100 virtual samples in total are generated to decrease the influence of the randomness caused by the meso-scale model in this case study. The results of the three basic statistical parameters, i.e., σ_d , μ_d and δ_d , are shown in Fig. 12(a), Fig. 12(b) and Fig. 12(c), respectively. From Fig. 12(a), it can be seen that σ_d increases with time in the early stage of the concrete carbonation process. However, when the carbonation front propagates into a typical position (about 0.02m in this case study), σ_d reaches a stable plateau where an almost constant value of σ_d can be found.

To provide a qualitative explanation on the above tendency of σ_d , the parameter called the distribution density of aggregates denoted as $D(y)$ is adopted here. The definition of this parameter is schematically shown in Fig. 13, and detailed information about the calculation method of this parameter can be found in Zheng *et al.* (2003). Generally, $D(y)$ is often used to present the local content of aggregates in a 2-D meso-scale model of the concrete. The result of $D(y)$ in this case study is plotted in Fig. 14. Due to the wall effect caused by the boundary of the meso-scale model, $D(y)$ near the boundary gradually increases from 0.0 to a stable value which is a little larger than the overall area fraction of aggregate, i.e., 0.5 in this case study.

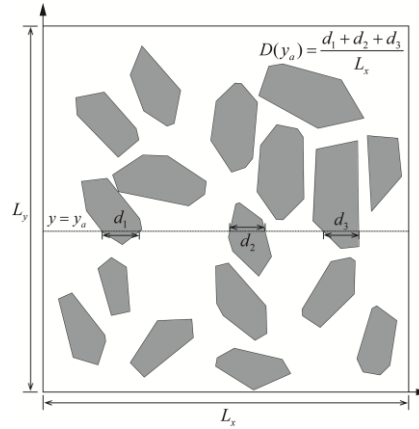


Fig. 13 Schematic illustration of $D(y)$

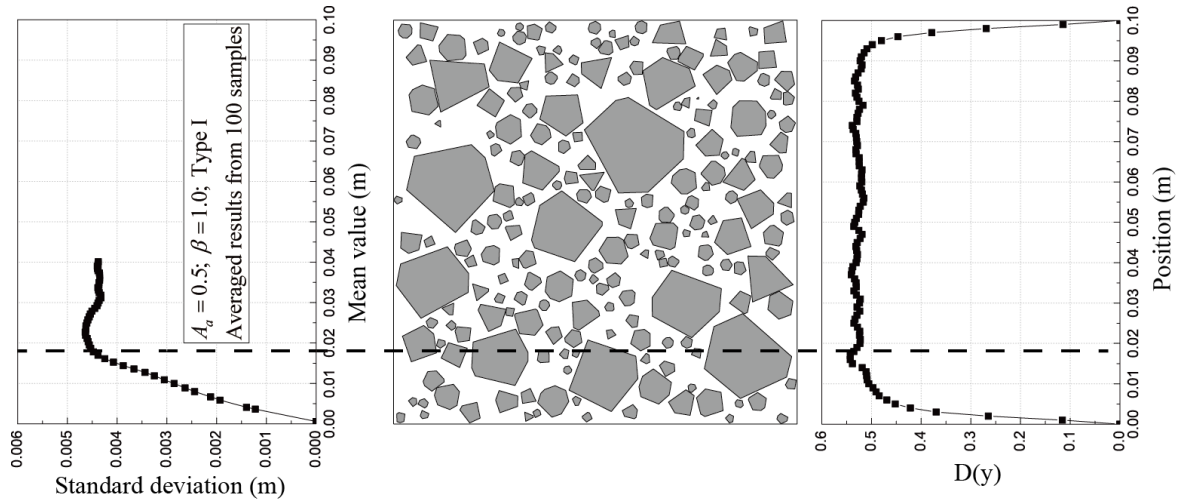
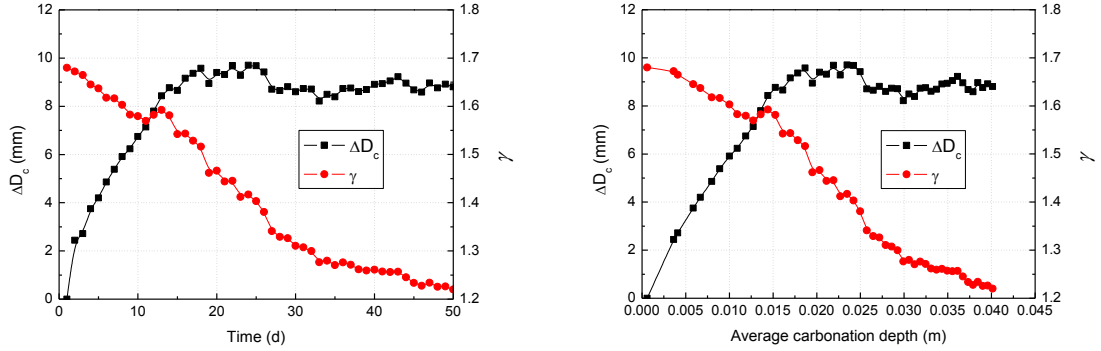


Fig. 14 A case study on qualitative relationship between σ_d and $D(y)$

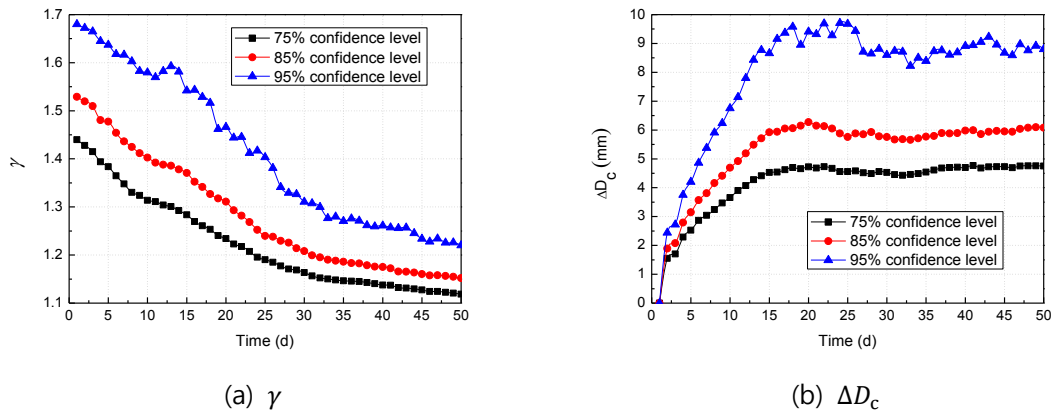
Since the spatial variability of the carbonation depth is mainly caused by the presence of the aggregates in the cement paste, σ_d should have a close relationship with $D(y)$ in the concrete. To provide a clearer explanation, the relationship between σ_d and μ_d of the carbonation depth is firstly plotted in the left part of Fig. 14, based on the results in Fig. 12(a) and Fig. 12(b). In the early stage of the concrete carbonation, the carbonation front only propagates in a small region near the surface of concrete, where $D(y)$ keeps increasing. Correspondingly, σ_d will also increase. However, when the carbonation front develops to a typical position inside the concrete, $D(y)$ reaches the ultimate stable value and stops increasing. Therefore, a stable value of σ_d will be also expected during the later carbonation process.

Based on Eqs. (14) and (16), an analysis on the parameters γ and ΔD_c is conducted. First of all, the parameter α is calculated according to the procedure proposed in Section 3.1. The result is shown in Fig. 12(d). It is interesting to find that the values of α present a slight oscillation around



(a) Relationship with time

(b) Relationship with average carbonation depth

Fig. 15 Analysis of parameters γ and ΔD_c for case study in Fig. 12(a) γ (b) ΔD_c Fig. 16 Effects of confidence level on γ and ΔD_c

2.0 during the carbonation process, which happens to be the value of α in the case of a normal distribution.

After α is calculated, γ and ΔD_c can then be determined according to Eqs. (14) and (16), respectively. The results are shown in Fig. 15. Since α is almost independent on the time according to the result in Fig. 12(d), γ shows the similar tendency with δ , while ΔD_c shows the similar tendency with σ_d . Therefore, ΔD_c also has an ultimate stable value, and this value can be reached provided that the concrete carbonation process can develop to a typical stage during the service life.

The effects of the confidence level on γ and ΔD_c are shown in Fig. 16. As expected, when the confidence level decreases, both of γ and ΔD_c decrease at the same time. However, when the confidence level as low as 75% is adopted, the value of ΔD_c in this case study is still larger than 4.0mm, which is not suitable to neglect in practical engineering even when the construction error is considered.

In the following sections, i.e., from Section 3.3 to 3.5, the effects of the meso-scale model of

the concrete on γ and ΔD_c are comprehensively studied. This study is of special importance to find the possible optimized aggregates or provide any useful suggestions on the proper mix proportion of concrete.

3.3 Effect of aggregate shape

In this paper, the shape of the aggregates is simplified as a polygon, whose geometry is more complicated to accurately describe than an ellipse used somewhere else (Zheng *et al.* 2012, Pan *et al.* 2014). However, according to the generation procedure of the polygons shown in Fig. 5, the aspect ratio is one of the major parameters controlling the shape of the aggregates. Therefore, we only focus our attention on studying the effect of the aspect ratio on the spatial variability of the carbonation depth in this paper.

Based on the statistical analysis of 10 types of aggregate particles by Wang *et al.* (2005), the aspect ratio of normal aggregates is around 2.0. Therefore, three different aspect ratios of aggregates, i.e., $\beta = 1.0, 2.0, 3.0$, are chosen. It should be pointed out that the aggregates with a too large aspect ratio are usually forbidden in the design codes in many countries, since the aggregates with a larger aspect ratio may be more likely to cause a severe stress concentration in the concrete. Therefore, it can be agreed that the range of the aspect ratio used here almost covers all the aggregates which can be really used in practice.

By following the same analysis procedure in Section 3.2, the results for these three different aspect ratios are obtained and shown in Fig. 17. Generally, the differences found among these cases do not have significant influence on the thickness of the concrete cover, especially after the consideration of the construction error. However, it is still valuable to point out that according to the results in Fig. 17, the degree of the spatial variability of the carbonation depth will increase

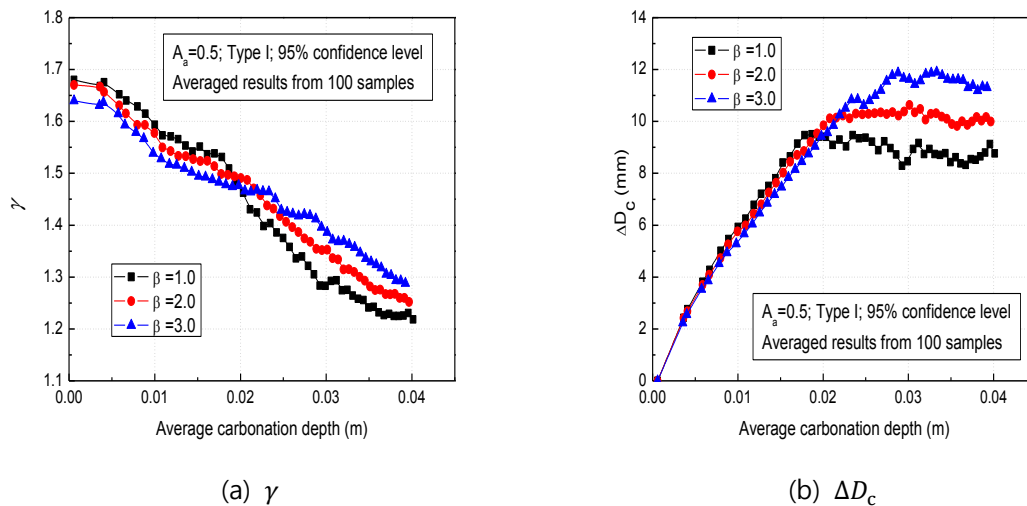


Fig. 17 Effect of aggregate aspect ratio on spatial variability of carbonation depth

with the aspect ratio of the aggregate. The main reason is that the concrete made from the aggregates with a larger aspect ratio usually have a more severe anisotropic material property from the probabilistic point of view. In this sense, although the aspect ratio of the aggregate has a quite limited effect on the thickness of the concrete cover, the aggregates with a smaller aspect ratio should still be preferred in practical engineering in that it can result in a smaller degree of the spatial variability of the carbonation depth.

3.4 Effect of aggregate content

Since the research in this paper is conducted in 2-D, the area fraction is used here to represent the aggregate content. Three different area fractions, i.e., $A_a = 0.5, 0.6, 0.7$, are adopted to study the effect of the aggregate content on the spatial variability of the carbonation depth. The results are shown in Fig. 18.

As can be seen, when the overall carbonation fronts propagate to the same position, both of γ and ΔD_c increase with the area fraction of the aggregates. This tendency again confirms the assumption that the spatial variability of the carbonation depth under the homogeneously exposed condition is mainly caused by the aggregates in the cement paste. When fewer aggregates exist in the cement paste, the possibility that the carbonation front meets the aggregates is smaller. Therefore, the carbonation front is less likely to be distorted by aggregates, which will result in a smaller degree of the spatial variability.

It can be also noticed that the average carbonation depth increases with the area fraction of the aggregates. For example, the ultimate average carbonation depth at the end of the virtual experiment ($t = 50d$) for $A_a = 0.7$ is 0.055m, while that for $A_a = 0.5$ is only 0.04m. This tendency is well supported by the experimental research by Papadakis *et al.* (1991b). However, it is not a

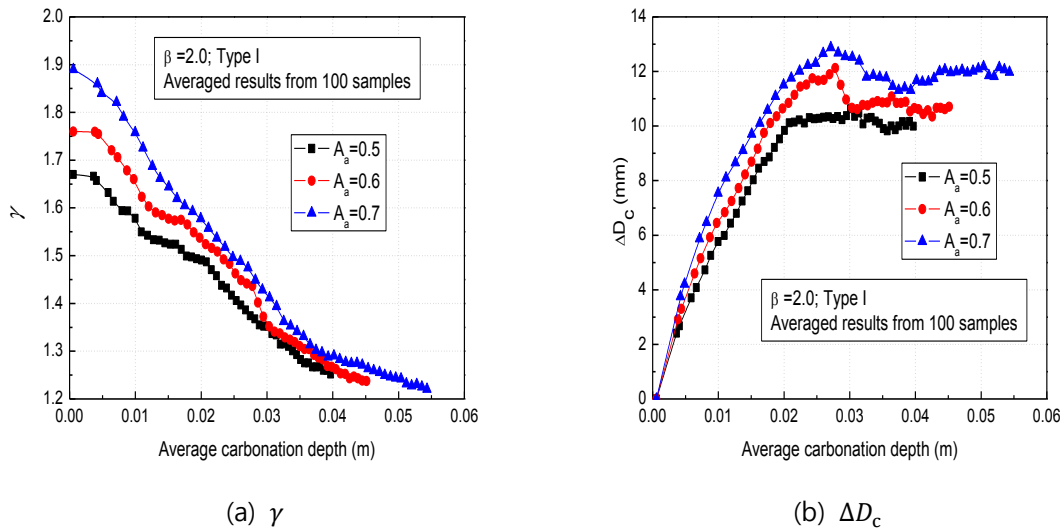


Fig. 18 Effect of aggregate content on spatial variability of carbonation depth

universal conclusion. At least three aspects should be considered to evaluate the effect of the aggregate content on the concrete carbonation.

(1) Generally speaking, when more aggregates are used in the concrete, less cement has to be used. Otherwise, there will not be enough water to ensure the workability of the concrete. Therefore, the capacity of the concrete to absorb CO_2 will decrease since the carbonation process can only take place in the cement paste.

(2) The aggregates cannot be penetrated by CO_2 , which means CO_2 has to move along the surface of the aggregates. In this way, the transport process of CO_2 is delayed.

(3) The ITZ around the aggregates has a higher porosity, and thus can provide a more convenient path for CO_2 to transport.

The overall effect of the aggregate content on the concrete carbonation is determined by all of these aspects. Therefore, it is very difficult to obtain any universal conclusions on this issue.

3.5 Effect of aggregate gradation

To study the effect of the aggregate gradation on the spatial variability of the carbonation depth, Two different types of aggregate gradations labelled as Type II and Type III shown in Fig. 19 are adopted. The main difference is that Type II has more small particles than type III

According to the results shown in Fig. 20, it can be seen that although the area fraction of the aggregates is kept the same, differences in γ and ΔD_c are still obtained for these two different aggregate gradations. It can be seen that γ and ΔD_c of Type II are smaller than those of Type III. This result suggests that the degree of the spatial variability of the carbonation depth should be mainly determined by the percentage of the large particles in the aggregate gradation. Small particles only have a quite limited effect on the spatial variability of the carbonation depth.

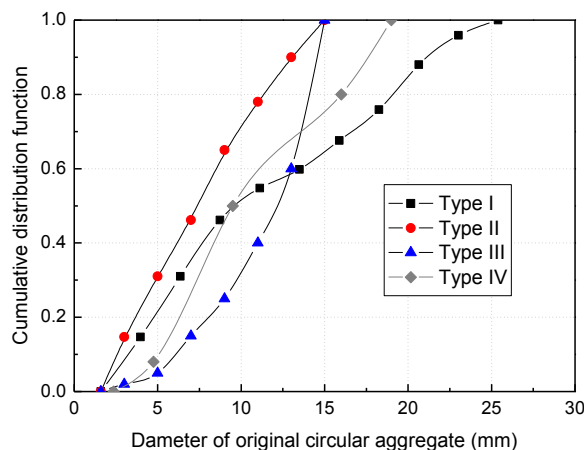


Fig. 19 Aggregate gradations used in this paper

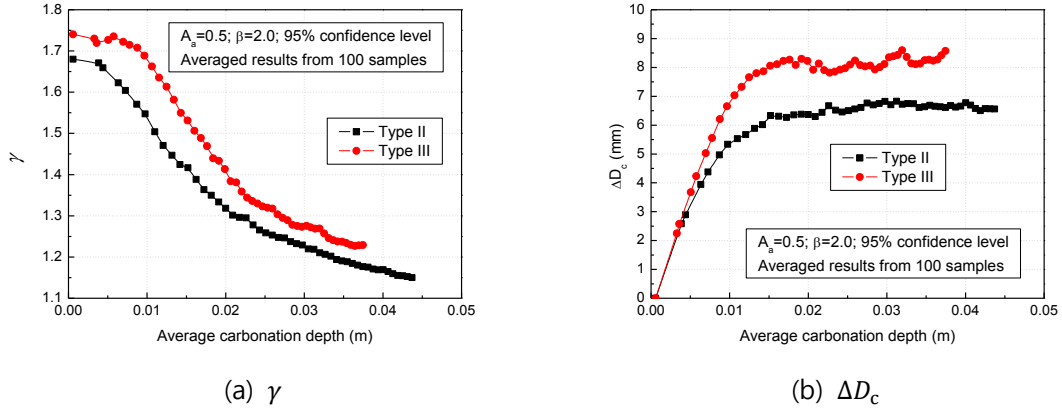


Fig. 20 Effect of aggregate gradation on spatial variability of carbonation depth

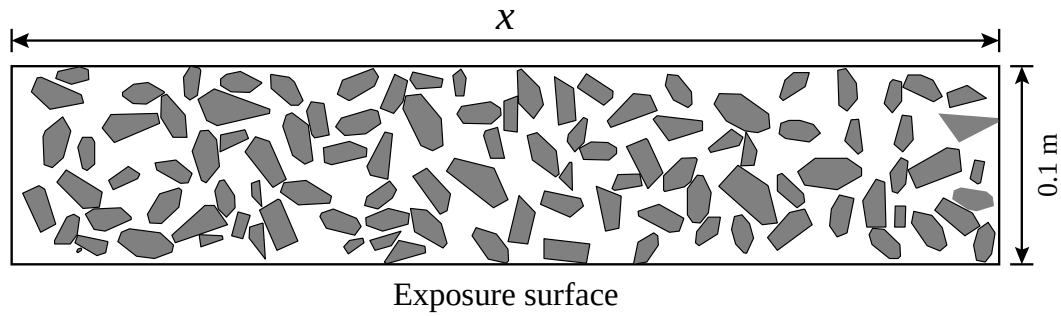


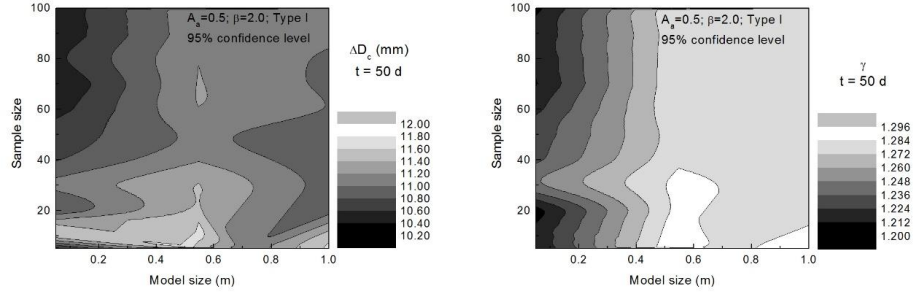
Fig. 21 Model used in analysis on size effect of meso-scale model

3.6 Proper sample size and model size used in simulation

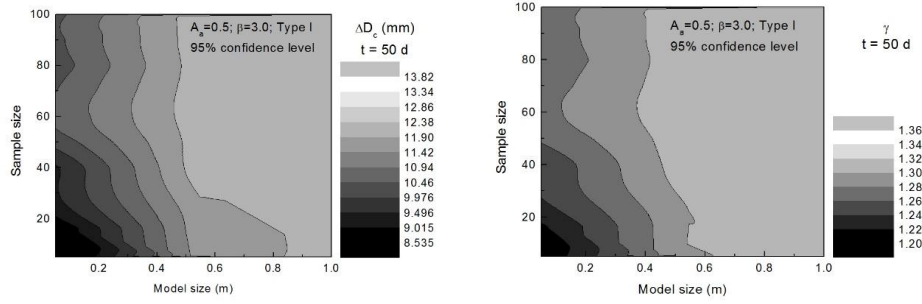
Before the above method is applied in practical engineering, the following two problems should be solved.

The first problem is how to determine the proper sample size. In the above analysis, the meso-scale model of the concrete is randomly generated. Therefore, the sample size should be large enough to eliminate the influence of the randomness caused by the meso-scale model on the analysis results. However, a larger sample size will absolutely result in more computational time, especially in a mesoscopic analysis.

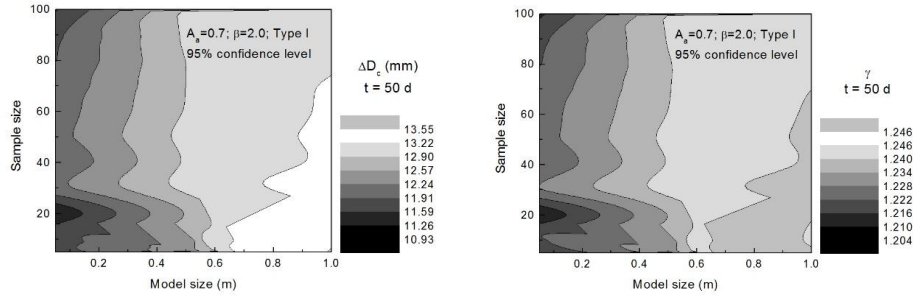
The second problem is how to determine the proper model size. In this above analysis, the size of the virtual samples can be just kept the same as that of the real samples used in experiments. However, when the analysis is conducted on the cross section of a real structure, it is impossible to choose the size of the entire cross section which is often several meters as the size of the meso-scale model. In most situations, only a small region near the exposure surface is cut from the cross section. The mesoscopic analysis is then performed in this region, and the obtained results at meso-scale can be passed back to macro-scale and assumed to be suitable in the whole cross



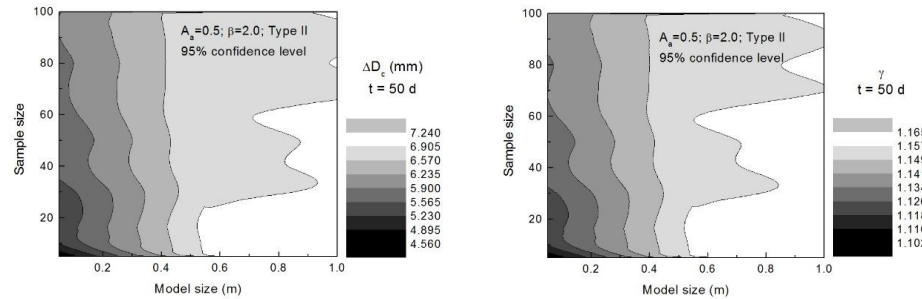
(a) Case 1



(b) Case 2



(c) Case 3



(d) Case 4

Fig. 22 Effects of sample size and model size on ΔD_c and γ

section. This is one of the traditional multi-scale modeling methods, and can be also called as “parameter passing method”. An important question in this method is how to determine the proper size of the small region. For the issue of the concrete carbonation, this region should be large enough to fully consider the spatial variability of the carbonation depth, while as small as possible to enhance the simulation efficiency.

To find the proper sample size and model size, the model used in the above virtual experimental research is redesigned. The modified model is shown in Fig. 21. Different sample sizes and different values of x are tried to find the effects of the sample size and model size on the parameters ΔD_c and γ . The results of four typical cases are shown in Fig. 22. Based on these results, the following conclusions can be made.

(1) Generally, the sample size of 100 is already large enough for these four cases. That is why this sample size is adopted in the above analysis. Furthermore, no obvious differences of the proper sample size can be found among these different cases. This result indicates that the parameters of the aggregates have ignorable influences on the proper sample size.

(2) When the sample size of 100 is used, the value of x should be larger than about 0.5m, in order to get a stable result of ΔD_c and γ . The parameters of the aggregates also do not show obvious influences on the proper model size.

4. Application

4.1 General information

To illustrate the influence of the spatial variability of the carbonation depth on the design of the thickness of the concrete cover, a simple supported bridge of a viaduct in an urban area is studied here. The girder of this bridge has a traditional T-shaped cross section, which is shown in Fig. 23. Based on the analysis in Section 3.6, a small region of $0.5 \text{ m} \times 0.1 \text{ m}$ is cut from the entire cross section to perform the mesoscopic simulation.

The annual average relative humidity in this urban area is about 63%. The concentration of CO_2 in the atmosphere is about 430ppm based on an in-situ test. However, since the bridge is expected to serve for 100 years, it is not suitable to neglect the possible increase of the CO_2 concentration due to the global warming effect during the service life. As a result, we do not use the result from the in-situ test here. A prediction model of the CO_2 concentration proposed by Stewart *et al.* (2011) is adopted instead. This model (labelled as A1FI in their paper) assumes a very rapid economic growth, which in our opinion should be the situation in China. In this case, the boundary value of $\text{CO}_2(\text{g})$ becomes a time-dependent variable, which is updated step by step in the solver. by fitting the data in Stewart *et al.* (2011), A formula is given in Eq. (17) to calculate the boundary value of $\text{CO}_2(\text{g})$ in each time step

$$[\text{CO}_2(\text{g})]_0 = 338.3 \cdot e^{0.01083 \cdot (\text{yr} - 2000)} \quad (17)$$

where yr here refers to the year (larger than 2000). The calculated $[\text{CO}_2(\text{g})]_0$ is in the unit of ppm.

Three mix proportions of the concrete shown in Table 2 are proposed. All these mix proportions can make the concrete reach a strength level of C40, which is required by the mechanical performance. All these mix proportions have the same aggregate gradation which is labelled as Type IV in Fig. 19. According to the w/c of these mix proportions, the initial concentrations of $\text{CH}(\text{s})$, as well as some other parameters used in the simulation are shown in Table 3.

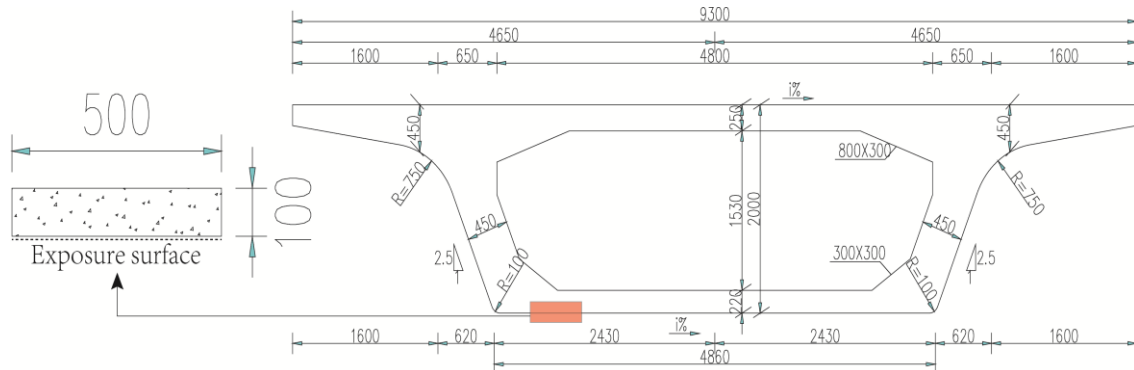


Fig. 23 Cross section of girder and model used in mesoscopic simulation (unit: mm)

Table 2 Mix proportions of concrete (unit: kg/m³)

No.	cement paste	water	sand	rock	superplasticizer
Mix 1	410	156	639	1242	3.08
Mix 2	363	156	655	1273	2.72
Mix 3	325	156	668	1298	2.44

Table 3 Parameters used in simulation of application case

Variable	Mix 1	Mix 2	Mix 3
ϕ_p^0	0.30	0.35	0.39
s^*	0.4654	0.4241	0.3865
$[\text{CH}(\text{s})]_0$	2280 mol/m ³	2130 mol/m ³	2000 mol/m ³

* The saturation degree is an average of the values for adsorption and desorption processes.

4.2 Meso-scale model

To get the aspect ratio of the real aggregates, a photo of some aggregates used in this project is taken. The outlines of these aggregates can be identified by some image processing software such as ImageJ. After the coordinates of the vertices of the aggregates are obtained, the aspect ratios of these aggregates can be calculated according to the definition shown in Eq. (1). The final aspect ratio used in the simulation is the average of these results. The whole process is shown in Fig. 24. Since the region used in the simulation is cut from the entire cross section, the wall effect should be eliminated on each boundary of the region except the exposure surface. Since an exposure surface should be included in the model, the periodic boundary condition (Bentz *et al.* 1992) is not suitable here. Thus, we extend the size of the region from 0.50m × 0.10m to 0.60m × 0.15m, including a so-called compensation area. The meso-scale model of the concrete is simulated in the larger region. However, only the original region is taken to perform the further analysis. This method is schematically shown in Fig. 25. The type of the boundary in this case study is also plotted in Fig. 25 (see Fig. 9 for the expressions of the boundary condition)

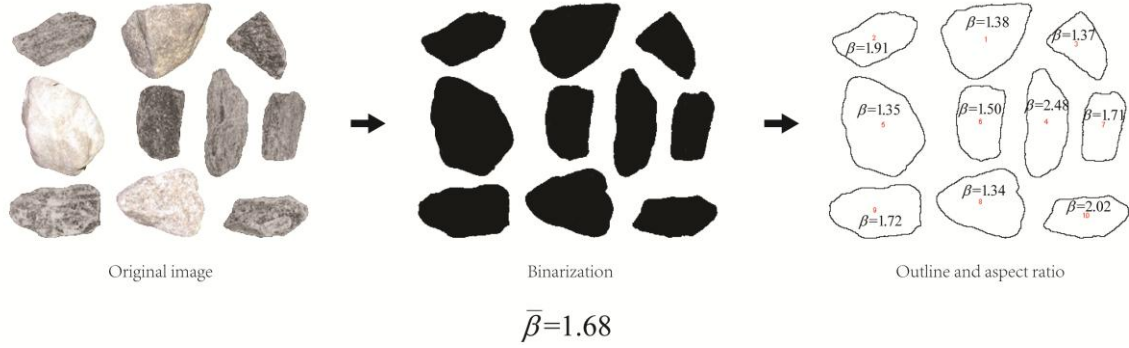


Fig. 24 Image-processing based method to determine aspect ratio of real aggregates

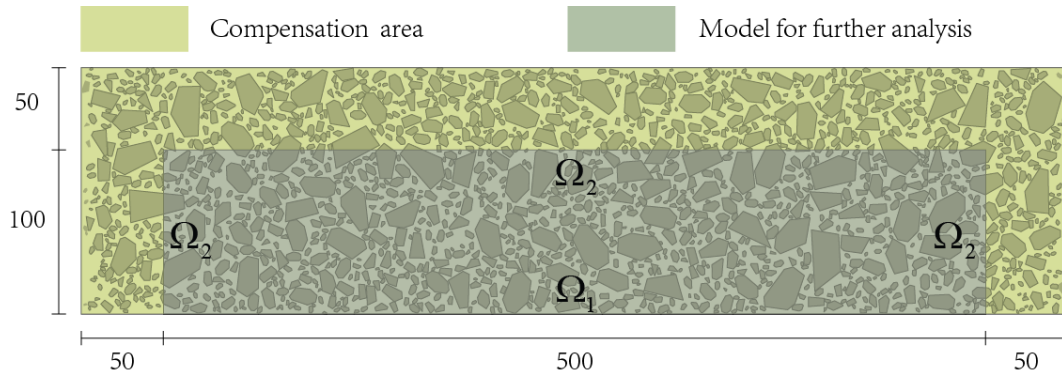


Fig. 25 A schematic of method to eliminate wall effect on distribution of aggregates (unit: mm)

4.3 Results and discussion

Based on the mix proportions of the concrete shown in Fig. 26 and the above meso-scale model, the parameters γ and ΔD_c in this case study can be obtained by following the same analysis procedure in Section 3.2. The results are shown in Fig. 26. According to these results, the thicknesses of the concrete cover for these three mix proportions can be calculated by Eqs. (13) and (14). The result is shown in Fig. 27.

As can be seen, Mix 1 should be adopted since the smallest thickness of the concrete cover is required by using this mix proportion. This result is reasonable in that Mix 1 has the lowest w/c among these three mix proportions. However, it should be pointed out that this is not a universal conclusion. With the consideration of the spatial variability of the concrete cover, a lower w/c cannot absolutely guarantee a smaller thickness of the concrete cover. The reason can be well explained by Eq. (14). Generally, a lower w/c can result in a smaller average carbonation depth due to the smaller porosity after cement hydration (Jennings and Tennis 1994) and the smaller effective diffusion coefficient of CO_2 (Papadakis *et al.* 1991b). However, according to the results shown in Fig. 17(a), Fig. 18(a) and Fig. 20(a), a smaller average carbonation depth can result in a larger partial factor. Since the final thickness of the concrete cover is calculated by multiplying the average carbonation depth with the partial factor. It is really difficult to provide any universal

conclusion whether the required thickness of the concrete cover decreases with the w/c after the consideration of the spatial variability of the concrete cover. Therefore, a case-by-case analysis is absolutely necessary to obtain the final suggestions on the mix proportion and the thickness of the concrete cover.

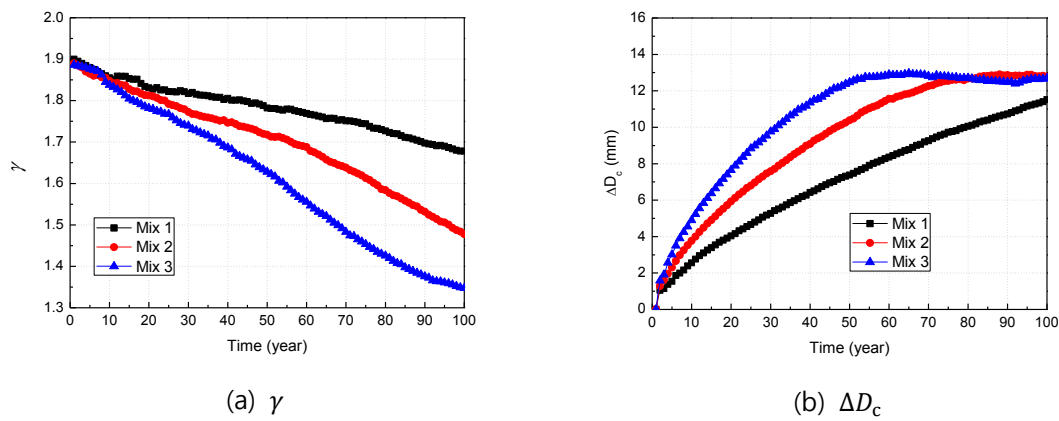


Fig. 26 γ and ΔD_c for different mix proportions

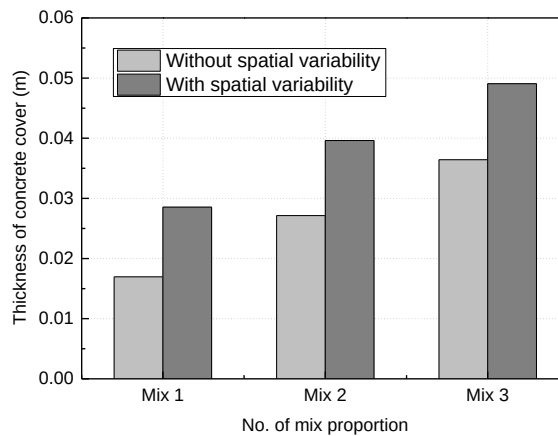


Fig. 27 Thicknesses of concrete cover for different mix proportions

5. Conclusions

The spatial variability of the carbonation depth is a normal phenomenon confirmed by the accelerated carbonation experiment. This phenomenon is mainly caused by the heterogeneity of the concrete at meso-scale. The effect of the mesoscopic structure of the concrete on the spatial variability of the carbonation depth is studied in this paper. Based on the results, the following conclusions can be made.

(1) A larger degree of the spatial variability will be obtained by using a) the aggregates with a larger aspect ratio; b) a larger aggregate content; c) the gradation which has more large particles.

(2) The effect of the spatial variability on the thickness of the concrete cover can be expressed as a partial factor. This partial factor decreases with the average carbonation depth.

(3) In order to eliminate the influence of the randomness caused by the meso-scale model of the concrete, the sample size should not be smaller than 100. When the sample size of 100 is used, the model size should be larger than 0.5 m to fully consider the spatial variability of the carbonation depth, in the case where the maximum size of aggregate is smaller than 26.5 mm.

(4) When the spatial variability is considered, a lower w/c cannot ensure a smaller thickness of the concrete cover. The proper mix proportion of the concrete and the thickness of the concrete cover should be determined based on case-by-case analysis.

Besides the above conclusions, this paper provides a deeper insight into the mesoscopic study of the concrete carbonation phenomenon, and forwards the current deterministic research into a probabilistic one in this field.

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